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(54) Title: MOISTURE-BARRIER EDIBLE COATING LAYER FOR FOODS

(57) Abstract: The invention relates to a moisture-barrier edible coating layer for foods. A particular advantage of this new coating layer is that it combines a high mechanical strength with improved moisture-barrier properties and heat stability. The invention also relates to foods to which such a moisture-barrier edible coating layer is applied or into which it is incorporated. These foods have, inter alia, the property that differences in moisture contents in various components of the food are maintained longer, so that storage for a longer period of time is possible without decrease in the flavor and the texture of the food.

Title: Moisture-barrier edible coating layer for foods

The present invention relates to a moisture-barrier edible coating layer for foods.

The preservability of foods is, in addition to being enhanced by factors preventing decay, enhanced by measures reducing the diffusion of moisture through the product. For instance, a moisture impermeable package around a loaf of bread prevents it from dehydrating rapidly.

The use of a coating layer having moisture-barrier properties is particularly advantageous for composite foods in which the water content in the various components is different and may not alter significantly during storage. A moisture-barrier coating layer in or on a food must contain only edible ingredients and be edible as a whole as well.

To realize this, a number of products, usually based on fat (i.e. chocolate and mayonnaise) are used to keep different components in composite foods having different moisture contents separated from each other. However, in practice, the moisture resistance of these so-called moisture-barriers is found to be too low, of too limited applicability or too short in duration. A moisture-barrier protective layer consisting of, for instance, chocolate can be affected by temperature. Mayonnaise can penetrate into the surrounding food or be incorporated therein, thereby causing, over time, the moisture-barrier protective layer to become thinner. For that reason, the moisture-barrier action and applicability of a number of existing, edible moisture-barriers are limited and improvement is desired.

Much research in the field of edible moisture-barriers, inter alia for coating tablets, has been carried out by the pharmaceutical industry. For instance, British patent application 756082 describes that the moisture sensibility of tablets can be reduced by mixing the moisture-sensitive powders with a solution of a prolamin in alcohol and then to process this into tablets.

US patent 3,477,864 also describes a coating layer for pharmaceutical applications, as on tablets. The coating layer contains as main component hydroxypropylmethyl cellulose. In all examples described aliphatic halides are used as solvent, which is inadmissible for use in foods. The action of the described coating layer is only shown for very short periods of time at relatively high humidity levels. Therefore, it cannot be expected that a coating layer as described in this document could be effective in foods, where protection from moisture transport is required for a long period of time.

The international patent application 97/46224 describes enteric coating layers for pharmaceutical preparations for oral administration. The coating layer is applied in several steps and contains as essential component alginic acid particles. These are particles of a charged carbohydrate polymer which is not soluble at low pH, but which is at a neutral or basic pH. With the aid of a binding agent, the alginic acid particles are anchored in a coating layer through bonding with an additional carbohydrate polymer (locust bean gum powder). At neutral pH, the coating layer dissolves in a short period of time when it is brought into contact with an aqueous medium, which is undesired for a moisture-barrier coating layer for foods.

In US patent 5,882,715 as well, an enteric coating layer for pharmaceutical preparations is described. The coating layer is applied in two or three steps. A first layer is applied for preventing the water activity of the pharmaceutical preparation from rising too much during the following step, which is the application of the pH-dependent alginate layer in the form of an aqueous solution. The eventual coating layer is not intended for use in composite foods in which the layer is constantly exposed to a high water activity.

Shellac, usually in combination with hydroxypropyl cellulose, is also an often used biopolymer in the application of a moisture-barrier coating on foods (see, inter alia, US 4,820,533). The combination of shellac with prolamin is used for this purpose as well (EP 0 090 559).

Prolamins, which can be obtained from the alcohol-soluble protein fraction from cereals, are widely used as film-forming component of a moisture-barrier and edible coating layer. One of the most important commercial prolamins is zein, the group of alcohol-soluble proteins from corn. However, at this moment, the commercial use of zein as component of moisture-barrier protective layers has an important drawback. In practice, moisture-barrier protective layers based on zein are found to be too brittle.

To reduce the breakage of protein containing moisture-barriers, a polyalcohol or a polyglycol can be added, as is described in EP 0 465 801. However, this solution has the drawback that the moisture-barrier properties of the coating layer are strongly restricted.

US patent 6,039,988 relates to a method wherein, successively, an edible oil layer and a membrane layer are applied to a baked substrate, such as a cookie or a waffle. The two layers then form a laminated structure. In practice, the manner of production of multi-layered coating layers is very laborious and expensive.

There is a strong need for edible coating layers with a better moisture-barrier action, with a longer life span, with broader applicability, with a higher temperature stability and with a greater elasticity. To a large extent, this need is motivated by the growth of the market of composite foods, including convenience food. In such foods, moisture-containing ingredients are combined with dry ingredients. Moisture migration or moisture diffusion occurring through the food decreases the flavor and the texture of the food, and in particular of the dry ingredients, in a rapid and considerable manner.

The invention contemplates obviating the above-mentioned problems. Surprisingly, it has been found that this is possible by preparing a coating layer based on prolamin in combination with a lipid and a salt of a fatty acid. In more detail, the invention provides a composition for a moisture-barrier edible coating layer for foods which composition comprises a prolamin, an alcohol, a lipid and a salt of a fatty acid. Such a coating layer has the

advantage that it has a very high moisture resistance and exhibits a high mechanical strength. In addition, the new coating layer is, to a large extent, impermeable to water and water vapor. Another important and surprising advantage of the invention is that the proposed moisture-barrier coating layer can endure being heated without appreciable damage to the coating layer. As a result, the coating layer can be used on, for example, dough still having to rise or dough which expands during the baking process.

As noted, the invention relates to a composition for a moisture-barrier edible coating layer for foods. The invention provides a method for preparing such a composition and a method for obtaining a moisture-barrier edible coating layer on foods. In particular, the invention describes a moisture-barrier edible coating layer with favorable properties which were not achieved in the past. The present moisture-barrier edible coating layer has as a particular advantage that it combines a high mechanical strength with improved moisture-barrier properties and heat stability. In addition, a coating layer according to the invention has a great integrity and, during preparation and storage of a food onto which it has been applied, does not considerably liquefy into its surroundings, but remains a separate phase and thus forms a stable barrier.

The invention also relates to foods onto which such a moisture-barrier edible coating layer has been applied or into which it has been incorporated. These foods have, inter alia, the property that differences in moisture content in different ingredients of the food are maintained longer, so that storage for a longer period of time is possible without decrease of the flavor and the texture of (the components of) the food.

Highly suitable for use in the compositions for the moisture-barrier-coating layer are prolamins. According to the invention, as prolamin, preferably gliadin from wheat or rye, zein from corn, hordein from barley, prolamin from millet or rice, avenin from oat and kafirin from sorghum are used.

Prolamins can, inter alia, be obtained by extracting the flour, obtained by grinding the grains, with a 70-100 percentage alcohol solution in water. To this, between 0 and 2 percents by weight of a reducing agent, for instance sulphite, can be added. The extraction can take place at a temperature between 20 and 80°C. For the extraction of kafirin and zein, preferably, a hot alcohol extraction is carried out. Alcohols which can be used for extraction are, inter alia, methanol, ethanol, propanol and butanol. Criteria for the selection of the alcohol used for the extraction is that it must efficiently separate a prolamin from the other protein fractions and that a prolamin of a desired purity is obtained.

A particular advantage of a prolamin for use in a moisture-barrier coating layer according to the invention is that it is poorly water-soluble. It has been found that particularly zein, kafirin and hordein can be advantageously used. Preferably, in the composition of a moisture-barrier coating layer, a prolamin is present in an amount of 10-90 percent by weight calculated on the weight of the composition.

When preparing a composition for a moisture-barrier coating layer according to the invention, preferably, a prolamin is dissolved in alcohol. During evaporation of the alcohol, the protein precipitates and after complete evaporation of the alcohol, a surface film consisting of a network structure of crystallized protein is left on the surface the solution was applied to. The selection of the alcohol used for the preparation of the composition and the application of the moisture-barrier coating layer onto the food can be different but also identical to the one used for the extraction of the prolamin. An important criterion for the selection of the alcohol used as a component for the composition of the moisture-barrier coating layer is that it must serve as a solvent for prolamin and that, after evaporation, it may not leave toxic or health damaging residues. Alcohols which can form part of the composition of the moisture-barrier coating layer are, inter alia, methanol, ethanol, propanol, isopropanol, butanol, pentanol or a combination thereof.

A large number of lipids are suitable to be used in a composition for a coating layer according to the invention. Lipid is understood to mean glycerol esterified with at least one, but preferably three fatty acids. If the lipid is liquid at room temperature, it is called an oil. If the lipid is solid at room temperature, a fat is involved. An advantage of the use of oil or fat in a composition for a moisture-barrier edible coating layer according to the invention is that, as a result, the elasticity of the eventual coating layer is increased. The presence of oil or fat reduces the brittle structure of the coating layer.

Preferably, in a composition according to the invention, oils are used such as soy oil, sunflower oil, groundnut oil, rapeseed oil, olive oil, palm oil and sesame oil. The skilled person can also modify the structure of the coating layer by adding fats with a melting temperature between 30 - 60°C. For instance palm fat, having a melting temperature of 45°C, or soy fat, having a melting temperature of 37°C, can be used. It will be clear that the use of combinations of oils and/or fats is also possible.

The selection of the type of oil or fat is mainly motivated by the intended use. For instance, for a food which still has to rise, and that, thereupon, will be baked in an oven, a different composition will be chosen than for a food which does not change in volume and is stored at 5°C.

Preferably, in a composition for a moisture-barrier coating layer according to the invention, the lipid is present in an amount of 0.1 – 50 percent by weight calculated on the weight of the composition.

An important component of a composition according to the invention is a salt of a fatty acid. An important advantage of this salt of a fatty acid is that it greatly strengthens the network in the moisture-barrier coating layer. Together with the prolamin, the salt of a fatty acid is dispersed in the alcohol and is preferably homogeneously distributed through the coating layer. Preferably, the salt is a bivalent metal salt, such as a magnesium salt, a zinc salt or a calcium salt. Preferably, the fatty acid is a higher fatty acid having a

length of 14 - 20 carbon atoms such as stearate, palmitate, oleate or arachidate. A desired property of the salt of a fatty acid is that it is poorly soluble in alcohol, water and alcohol-water mixtures. Preferably, the melting temperature of the salt of a fatty acid is over 100°C to guarantee the stability of the coating layer during a baking process. The degree of saturation or unsaturation of the fatty acid can be suitably selected depending on the desired properties. Preferably, in a composition for a moisture-barrier coating layer according to the invention, the salt of the fatty acid is present in an amount of 0.1 – 50 percent by weight, calculated on the weight of the composition. Preferably, magnesium stearate is used. Magnesium stearate strongly reduces the permeability of the moisture-barrier coating layer to water and water vapor.

A component which can be used with additional advantage in a composition according to the invention is an emulsifying fatty acid. The presence of an emulsifying fatty acid is particularly desired when a stable, mechanically strong coating layer has to be obtained. A particular advantage of the use of an emulsifying fatty acid is that with this, a highly homogenous coating layer is obtained. However, not in all cases is the presence of an emulsifying fatty acid essential.

Preferably, in a composition for a moisture-barrier coating layer according to the invention, the emulsifying fatty acid is present in an amount of 0.01 – 10 percent by weight calculated on the weight of the composition. Preferably, a fatty acid having a chain length of 6 – 12 carbon atoms is selected, such as hexanoic acid and lauric acid. There is a great preference for a fatty acid having a chain length of 10 –12 carbon atoms. Fatty acids having a shorter chain length can produce an unpleasant aroma, although this has no effect on the moisture-barrier properties of the coating layer.

In a preferred embodiment, the composition for a moisture-barrier coating layer according to the invention mainly consists of only the above-mentioned components, i.e. a prolamin, an alcohol, a lipid, a salt of a fatty acid,

and, optionally, an emulsifying fatty acid. In this context, the expression "mainly consist of only" is understood to mean that, in addition to the components mentioned, inevitable impurities can be present. An example thereof is a small amount of water, for instance in the alcohol.

For the preparation of a composition for a moisture-barrier coating layer, according to the invention, a prolamin and a salt of a fatty acid are preferably mixed dry. This can for instance be done with a spatula, but different manners are also possible. It is desired that the salt of a fatty acid is mixed in dry form with prolamin because then, a particularly homogenous coating layer is obtained. The mixture of a prolamin and salt of a fatty acid is dispersed in an alcohol by adding the alcohol to the mixture of the prolamin and salt of a fatty acid. While stirring continuously, the thus obtained dispersion can, optionally, be heated to 80°C. Preferably, the temperature is increased to 60°C to obtain as complete a dissolution of the zein as possible. During the preparation of the composition a certain amount of alcohol will evaporate. Preferably, the loss of alcohol is replenished during and at the end of the preparation. Finally, to the mixture, optionally, an emulsifying fatty acid and, at choice, oil or molten fat are slowly added while stirring.

The thus obtained composition can be applied to a food by means of spreading, sprinkling, spraying, atomizing, dipping, brushing and/or rolling. [illegible] at a temperature of about 60°C, depending on the melting point of the oil or the fat used as component. Preferably, the composition applied to the food is then left to dry. This can be done at room temperature or during a baking process. Preferably, the alcohol is substantially completely evaporated from the composition. After evaporation of the alcohol, a moisture-barrier edible coating layer having the above-described properties is left on the food.

The minimum thickness of the coating layer is determined by the grain-size of the salt of a fatty acid used in the composition. Depending on the use, a thicker or thinner coating layer will be desired. A minimal thickness of

~50 micrometers is possible. The maximally attainable thickness is a few millimeters. When using very finely ground powders of salt of a fatty acid, coating layers with a thickness of less than ~50 micrometers are possible. Preferably, the coating layer has a thickness between 50 and 500 micrometers.

In a preferred embodiment, the coating layer consists of one single layer, which is preferably of homogonous composition.

Foods on which or in which the moisture-barrier edible coating layer can preferably be used are composite foods in which the regulation of moisture migration is desired so as to maintain the product quality. Often, the moisture-barrier coating layer will be applied between different layers in the food or between different parts. It is possible, in advance, to provide moisture-sensitive parts of a food with a moisture-barrier coating layer and, thereupon, to incorporate it into the food. For instance, on the bottom of a ready-made pizza, a coating layer according to the invention can be applied resulting in the tomato sauce applied thereon not soaking the bottom. Also, in a different case, the bread or the cheese of a prepackaged bread roll can be provided with a coating layer according to the invention so that bread and cheese are no longer sensitive to moisture coming from vegetables that might be present.

Foods onto which or in which a moisture-barrier edible coating layer according to the invention can be applied are, inter alia

- pastry and cake with a stuffing, including flan, pizza, quiche, waffle or pancake;
- bread with a stuffing including sausage roll and almond-paste pastry;
- fruits, chocolates, nuts, muesli, cereals, cornflakes, candy, waffles, or (pan)cakes processed in custard, ice cream, milk, icing, soft cheese, pudding, syrup, yogurt or other liquid products;
- ready-made sandwiches and toast, including prepackaged bread rolls and sandwiches;
- stuffed products such as Evergreen™, Switch™ or muesli-bars.

Further, the moisture-barrier edible coating layer can be used for preventing uptake of moisture from the air by a composite or non-composite food. Here, cookies and other products with a low water activity are involved, or products destined for consumption in a tropical climate. Also, the moisture-barrier edible coating layer can be used in or be applied to products whose preservability during long transport has to be improved. Further, the coating layer can be used on or in products in which regulation of moisture migration is desired for maintaining or even improving the specific characteristics of this product, such as, for instance, crunchy muesli. Non-edible parts of composite foods such as packaging material, separating paper, napkins or chopsticks and the like can also be provided with a coating layer according to the invention.

The invention will presently be further elucidated with reference to the following examples which should be understood to be non-limitative.

Example 1. Moisture-barrier edible coating layer (coating) for pizza.

A moisture-barrier edible coating layer according to the invention was prepared to obstruct the moisture coming from the stuffing of a fresh, cooled pizza during the storage at 4°C. So as not to be restricted to specific ingredients on a pizza, the water activity (Aw) of different ingredients of a fresh, cooled pizza is measured. The Aw was determined with standard methodologies on an Aqualab AQ CXII water activity meter. The highest Aw measured was 1. In the test, a starch gel with this Aw is used. If this gel is laid on the pizza bottom, a water film is formed beneath the gel.

The composition of the coating layer is represented in Table 1. The composition of the pizza dough is represented in Table 2 and the preparation process is represented in Tables 3 and 4. In this example, the coating layer is applied with a brush on the raw dough. Therefore, the coating layer undergoes a baking step.

Table 1: Zein/lauric acid/oil-coating for fresh, cooled pizza.

Ingredient	weight (g)	Details
Zein	10.0	Fluka, Zein of corn, Mr 25,000-29,000
Mg-stearate	3.0	Fluka, pract., 3.8-5% Mg
Ethanol	15.0	96%
Lauric acid	0.5	Aldrich, purity 95.5%
Oil/fat	3.0	Soy oil

Table 2: Recipe pizza dough.

Ingredient	weight (g)
Flour	2000
Water	1020
Baking powder (ammonium bicarbonate, $(NH_4)HCO_3$)	40
Acid sodium pyrophosphate ($Na_2H_2P_2O_7$)	20
Salt (NaCl)	40
Fat (lard)	100
Potassium sorbate	8
Lactic acid	8
Cysteine solution (1 g/l)	40
Total dough mass	3276

The dough is kneaded in a spiral kneader Sinmag SM10. Further process steps are described in the following Table (Table 3).

Table 3: The process

Process step	Details
Dry mixing ingredients	1 min.
Adding water	temperature 0 °C
Slow mixing (spiral kneader, Sinmag SM10)	4 min.
Rapid kneading (spiral kneader, Sinmag SM10)	10 min. to 20°C
Rolling out (Rondo Seewer sheeter)	2.20 mm (setting)
Rising/Resting	10 min. at 10°C
Cutting out round pizza shape	

Baking is carried out in a Rototherm industrial oven according to conditions represented in the following table (Table 4).

Table 4: The baking process: Settings Rototherm industrial oven.

	Time		valve*
Steam	2 min.	1 liter	
Baking	5 min.	210 °C	
Baking	1 min.	210 °C	100%

*Valve position of Rototherm industrial oven.

After baking, the starch gel was laid on the pizza bottom and the product with coating layer and without coating layer (control) was stored at 10°C for a period of 25 days. After 25 days, the Aw of the pizza crust with coating layer had increased from 0.93 to 0.96. This increase still results in a pizza with a crunchy crust. According to the experts, a pizza crust with an Aw well above 0.97 is no longer crispy/crunchy. With the test pizza (without coating layer), after one day of storing, the pizza crust has already adopted the Aw of the gel. With the coating layer described, the quality of the pizza can be improved and the preservability can be considerably prolonged.

Example 2. Coating layer (coating) for biscuit.

The coating described in Table 5 is used to protect biscuit when stored at 20°C and 80% relative air humidity (RH).

Table 5: Zein/oil-coating for coating biscuit

Ingredient	weight (g)	Details
Zein	5.0	Fluka, Zein of corn, Mr 25,000-29,000
Mg-stearate	3.0	Fluka, pract., 3.8-5% Mg
Ethanol	7.5	96%
Oil/Fat	16.0	(75% soy fat 45* and 25% palm fat 37*)

* Melting point in °C of the fat and/or the oil.

The coating is applied onto biscuits named Knappertjes™ (Verkade, Zaandam, NL) by means of brushing. After application of the coating, the biscuit is dried at 160°C for 10 minutes. Thereupon, these biscuits and control biscuits (uncoated Knappertjes™, having, for that matter, been subjected to the same treatment) were stored in a climate chamber at 20°C and 80% RH. During the storage period, the increase in weight was measured. As shown in Table 6, the water uptake of the coated biscuit was considerably lower than that of an uncoated control biscuit.

Table 6: Increase of weight of coated and uncoated Knappertjes™ during storage at 20°C and 80% RV.

Increase in weight over time	3 hours	19 hours	115 hours	214 hours
Control biscuit	0.23 g	0.60 g	0.74 g	0.78 g
Zein/oil-coating coated biscuit	0.04 g	0.16 g	0.27 g	0.55 g

Example 3. Coating of coffee cookies for desserts

A coating having a composition as described in Table 7 is used to coat coffee cookies (Albert Heijn, Zaandam, NL).

Table 7: Zein/oil-coating for coating coffee cookies.

Ingredient	weight (g)	Details
Zein	29	Fluka, Zein of corn, Mr 25,000-29,000
Mg-stearate	9	Fluka, pract., 3.8-5% Mg
Ethanol	43	96%
Stearic acid	2	
Oil/Fat	17	Soy oil

After application of the coating, the coffee cookies are dried at 160°C for 10 minutes. These cookies and control cookies (uncoated coffee cookies having been subjected to the same treatment) are stored in custard. During the storage period, the Aw increase of the product is measured. As appears from Table 8, the employed coating gives a considerable increase in the resistance against moisture uptake.

Table 8. Increase in Aw of coffee cookies during storage in custard

Aw increase in time	0 hour	22 hours	46 hours
Control cookie	0.28	0.51	0.78
Zein-coating coated cookies	0.20	0.39	0.57

Claims

1. A composition for a moisture-barrier edible coating layer for foods, which composition comprises a prolamin, an alcohol, a lipid and a salt of a fatty acid.

2. A composition according to claim 1, wherein the salt of a fatty acid has a chain length of 14 – 20 carbon atoms.

3. A composition according to any one of the preceding claims, wherein the salt of a fatty acid is magnesium stearate.:

4. A composition according to any one of the preceding claims, also comprising an emulsifying fatty acid having a chain length of 10 – 12 carbon atoms.

5. A composition according to claim 4, wherein the emulsifying fatty acid is lauric acid.

6. A composition according to any one of the preceding claims, wherein the prolamin is a gliadin, a zein, a hordein, an avenin, a kafirin or a combination thereof.

7. A composition according to any one of the preceding claims, wherein the prolamin is present in an amount of 10 – 90 percent by weight calculated on the weight of the composition.

8. A composition according to any one of the preceding claims, wherein the prolamin is a zein.

9. A composition according to any one of the preceding claims, wherein the lipid is soy oil, sunflower oil, groundnut oil, rapeseed oil, olive oil, sesame oil, palm fat or soy fat or a combination thereof.

10. A composition according to any one of the preceding claims, wherein the salt of a fatty acid is present in an amount of 0.1 – 50 percent by weight calculated on the weight of the composition.

11. A composition according to any one of the preceding claims, wherein the lipid is present in an amount of 0.1 – 50 percent by weight calculated on the weight of the composition.

12. A composition according to any one of claims 4 – 11, wherein the emulsifying fatty acid is present in an amount of 0.01 – 10 percent by weight calculated on the weight of the composition.

13. A method for preparing a composition according to any one of the preceding claims, wherein a prolamin and a salt of a fatty acid are mixed dry and dispersed in an alcohol for obtaining a dispersion which is if required heated and to which a lipid is added.

14. A method according to claim 13, wherein the alcohol is methanol, ethanol, propanol, isopropanol, butanol or pentanol or a combination thereof.

15. A method for applying a moisture-barrier edible coating layer onto a food, wherein a composition according to claims 1 – 12 is applied onto the food.

16. A method according to claim 15, wherein the composition is applied by means of spreading, sprinkling, spraying, atomizing, dipping, brushing and/or rolling.

17. A method according to claim 15 or 16, wherein the coating layer is dried.

18. A moisture-barrier edible coating layer for foods comprising a prolamin, a lipid and salt of a fatty acid.

19. A food comprising a coating layer according to claim 18.

20. A food according to claim 19 which is a composite food.

INTERNATIONAL SEARCH REPORT

PCT/NL 02/00600

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 A23P1/08 A21D13/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 A23P

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
WPI Data, PAJ, EPO-Internal, FSTA

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 477 864 A (TUJI YOJI) 11 November 1969 (1969-11-11) cited in the application example 4	1-3,6-8, 10,11, 15-20
X	WO 97 46224 A (BIFODAN AS ;BRANDSBORG ERIK (DK); MARCUSSEN JOERN (DK)) 11 December 1997 (1997-12-11) cited in the application examples	18-20
X	US 6 039 988 A (MONTE WOODROW C) 21 March 2000 (2000-03-21) cited in the application examples 22,23	1,2,6-8, 10,11, 15-20

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
13 January 2003	22/01/2003

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Vuillamy, V

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